

This Page Is Inserted by IFW Operations
and is not a part of the Official Record

BEST AVAILABLE IMAGES

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images may include (but are not limited to):

- BLACK BORDERS
- TEXT CUT OFF AT TOP, BOTTOM OR SIDES
- FADED TEXT
- ILLEGIBLE TEXT
- SKEWED/SLANTED IMAGES
- COLORED PHOTOS
- BLACK OR VERY BLACK AND WHITE DARK PHOTOS
- GRAY SCALE DOCUMENTS

IMAGES ARE BEST AVAILABLE COPY.

**As rescanning documents *will not* correct images,
please do not report the images to the
Image Problem Mailbox.**

Please amend the specification without prejudice, without admission, without surrender of subject matter, and without any intention of creating any estoppel as to equivalents, as follows:

A¹
Page 1, line 1, insert - This application is a Continuation-in-Part of PCT/GB00/00013, filed January 5, 2000, designating the U.S., published July 29, 2000 as WO 00/42234 and claiming priority from GB 9900955.7 filed January 15, 1999. All of the above-mentioned applications, as well as all documents cited herein and documents referenced or cited in documents cited herein, are hereby incorporated herein by reference. - -

Page 9, please cancel paragraphs 9 and 10.

Page 11, please cancel the last paragraph.

Page 12, please cancel the first paragraph.

Page 12, paragraph 2, please amend as follows: - Figure 7 illustrates an X-ray diffraction pattern of a BaZrO₃ film as fabricated by Example 1;--

Page 12, paragraph 3, please amend as follows: - Figures 8(a) and (b) illustrate surface and cross-sectional SEM micrographs of a CdS film as fabricated by Example 2; and --

Page 12, paragraph 4, please amend as follows: - Figures 9(a) and (b) illustrate surface and cross-sectional SEM micrographs of a porous SiO₂ film as fabricated by Example 3.--

Page 18, please cancel lines 5 through 32.

Please cancel pages 19 through 21.

Page 22, please cancel lines 1 through 13.

Page 22 to 23, last paragraph, please amend as follows: - A non-aqueous precursor solution for the deposition of a BaZrO₃ film was first prepared as follows. Barium metal (as supplied by Aldrich) was completely dissolved in a volume of 2-methoxyethanol (as supplied by Aldrich) by stirring at room temperature to form a barium alkoxide solution. A stoichiometric

A⁵
059475050
amount of zirconium n-propoxide, a 70 wt% solution in n-propanol (as supplied by Aldrich), was then added to the barium methoxyoxide solution and refluxed at 124 °C, the boiling point of 2-methoxyethanol, for five hours. Then, a volume of 2-methoxyethanol was added to the refluxed solution to provide a 0.05 M precursor solution. Using the apparatus of the first-described embodiment and the so-prepared solution, a BaZrO₃ film was deposited on a silver substrate 5, with a substrate temperature of 600 °C, a substrate 5 to nozzle unit 11 distance of 30 mm, an electric field voltage of 10 kV, the piezoelectric transducer 43 of the aerosol generator 25 being operated at a frequency of 1.7 MHz and power of 50 W, and nitrogen being supplied at 30 ml per minute as the carrier gas. Nitrogen was used as the carrier gas to minimise the reaction between the barium and carbon dioxide in the air. The resulting film, formed in a single run without the need for any post-deposition heat treatment, was a crystalline BaZrO₃ film as characterized by the X-ray diffraction pattern illustrated in Figure 7.

A⁶
059475050
Page 23, the first full paragraph, please amend as follow: - A 0.01 M aqueous precursor solution for the deposition of a CdS film was first prepared using cadmium chloride and thiourea. Using the apparatus of the second-described embodiment and the so-prepared solution, a CdS film was deposited on a glass substrate 105, with a substrate temperature of 450 °C, a substrate 105 to nozzle unit 111 distance of 20 mm, an electric field voltage of 10 kV, the piezoelectric transducer 143 of the aerosol generator 125 being operated at a frequency of 1.7 MHz and power of 50 W, a deposition time of five minutes, and air being supplied at 50 ml per minute as the carrier gas. The resulting film, formed in a single run without the need for any post-deposition heat treatment, was a dense, crystalline CdS film having a thickness of about 1 µm, with a columnar structure and a smooth and uniform surface. SEM micrographs of the resulting film are illustrated in Figures 8(a) and (b).

Page 23, the second full paragraph, please amend as follows: A colloidal silica solution

(Ludox™, as supplied by DuPont) was diluted with distilled water to prepare an aqueous precursor solution having a concentration of 0.1 g/ml for the deposition of a SiO₂ film. Using the apparatus of the second-described embodiment and the so-prepared solution, a SiO₂ film was deposited on a glass substrate 105, with a substrate temperature of 200 °C, a substrate 105 to nozzle unit 111 distance of 20 mm, an electric field voltage of 10 kV, the piezoelectric transducer 143 of the aerosol generator 125 being operated at a frequency of 1.7 MHz and power of 20 W, a deposition time of one minute, and air being supplied at 50 ml per minute as the carrier gas. The resulting film, formed in a single run without the need for any post-deposition heat treatment, was a porous SiO₂ film with a reticular structure. SEM micrographs of the resulting film are illustrated in Figures 11(a) and (b).

Page 24, first paragraph, please amend as follows: Finally, it will be understood that the

present invention has been described in its preferred embodiments and can be modified in many different ways within the scope of the invention as defined by the appended claims. For example, in coating substrates 5, 105 of large area or complex geometric shape, the nozzle units 11, 111 could be modified to include a plurality of outlet ports 18, 118 or the film deposition apparatus could be modified to include a plurality of nozzle units 11, 111.

Page 24, after the first paragraph, please insert:

--The invention will now be further described by the following numbered paragraphs.

1. A method of depositing material, preferably a film, on a substrate, comprising the steps of:
providing a substrate;
heating the substrate;

generating an aerosol comprising droplets of a material solution;
providing a nozzle unit for delivering the aerosol to the substrate, the nozzle unit
including at least one outlet through which a directed flow of the aerosol is
delivered and at least one electrode;
charging the aerosol droplets with a positive or negative charge;
providing a flow of the aerosol through the nozzle unit so as to deliver a directed
flow of the aerosol from the at least one outlet; and
generating an electric field between the substrate and the at least one electrode
such that the directed aerosol flow is attracted towards the substrate.

Sub
ri
uncle

2. The method of paragraph 1, wherein the substrate is heated to a temperature of
less than about 1050 °C, preferably less than about 800 °C.

SUB
B₂

The method of paragraph 1 or 2, wherein the substrate is heated during
deposition,

4. The method of paragraph 3, wherein the thermal environment is such as to
maintain a decreasing temperature gradient in a direction away from the substrate
towards the nozzle unit.

5. The method of any of paragraphs 1 to 4, wherein the material solution is an
aqueous solution.

6. The method of any of paragraphs 1 to 4, wherein the material solution is a non-
aqueous solution.

7. The method of any of paragraphs 1 to 6, wherein the aerosol droplets are at least
partially charged prior to exiting the at least one outlet.

8. The method of paragraph 7, wherein the aerosol droplets are charged prior to

A⁹
TOTAL 674556-2001

exiting the at least one outlet.

9. The method of any of paragraphs 1 to 7, wherein the aerosol droplets are at least partially charged after exiting the at least one outlet.
10. The method of any of paragraphs 1 to 9, wherein the aerosol droplets are charged by the at least one electrode.
11. The method of any of paragraphs 1 to 10, wherein the at least one electrode is disposed at least partially in each aerosol flow.
12. The method of any of paragraphs 1 to 11, wherein the at least one electrode extends upstream of the at least one outlet.
13. The method of any of paragraphs 1 to 12, wherein the at least one electrode comprises an elongate element.
14. The method of any of paragraphs 1 to 13, wherein the distal end of the at least one electrode is located at substantially the centre of the at least one outlet.
15. The method of any of paragraphs 1 to 14, wherein the distal end of the at least one electrode includes a single tip.
16. The method of any of paragraphs 1 to 14, wherein the distal end of the at least one electrode includes a plurality of tips.
17. The method of any of paragraphs 1 to 16, wherein the nozzle unit includes a tubular section upstream of each outlet.
18. The method of paragraph 17, wherein the tubular section is an elongate section.
19. The method of paragraph 17 or 18, wherein the tubular section is a linear section.
20. The method of any of paragraphs 17 to 19, wherein the tubular section is substantially cylindrical.

21. The method of any of paragraphs 17 to 20, wherein the at least one electrode extends substantially entirely through the associated tubular section.
22. The method of any of paragraphs 17 to 21, wherein the at least one electrode extends substantially along the central axis of the associated tubular section.
23. The method of any of paragraphs 1 to 22, wherein at least the inner surface of the tubular section is composed of an insulating material.
24. The method of any of paragraphs 1 to 23, wherein the aerosol flow is provided by entraining the aerosol in a flow of a carrier gas fed to the nozzle unit.
25. The method of any of paragraphs 1 to 23, wherein the aerosol flow is provided by applying a reduced pressure to the at least one outlet so as to entrain the aerosol in a flow of a carrier gas drawn through the nozzle unit.
26. The method of paragraph 24 or 25, wherein the carrier gas is a gas reactive to the material solution.
27. The method of paragraph 24 or 25, wherein the carrier gas is a gas non-reactive to the material solution.
28. The method of any of paragraphs 24 to 27 when appendant upon paragraph 4, wherein the flow of the carrier gas is provided such as to maintain the decreasing, temperature gradient.
29. The method of any of paragraphs 1 to 28, wherein the aerosol is delivered to the substrate such as to achieve a film growth rate of at least 0.2 um per minute, preferably at least 1 um per minute, more preferably at least 2 um per minute.
30. The method of any of paragraphs 1 to 29, wherein the flow rate through the at least one outlet is at least 5 ml per minute, preferably at least 50 ml per minute.

31. The method of any of paragraphs 1 to 30, wherein the nozzle unit is configured such that the directed aerosol flow from the at least one outlet is directed upwards, preferably substantially vertically upwards.
32. The method of any of paragraphs 1 to 31, wherein the nozzle unit includes a perforated member upstream of the at least one outlet.
33. The method of any of paragraphs 1 to 32, wherein the applied voltage is less than about 35 kV, preferably less than about 20 kV.
34. The method of any of paragraphs 1 to 33, wherein the distance between the at least one outlet and the substrate is less than about 100 mm, preferably less than about 50 mm.
35. The method of any of paragraphs 1 to 34, wherein the substrate is held stationary relative to the nozzle unit.
36. The method of any of paragraphs 1 to 34, further comprising the step of moving the nozzle unit relative to the substrate.
37. The method of paragraph 36, wherein the substrate is rotated, tilted and/or translated relative to the nozzle unit.
38. The method of any of paragraphs 1 to 37, when performed at atmospheric pressure.
39. The method of any of paragraphs 1 to 37, when performed below atmospheric pressure.
40. The method of any of paragraphs 1 to 37, when performed above atmospheric pressure.

090917.071301
A9

41. An apparatus for depositing material, preferably a film, on a substrate, comprising:
- a substrate holder for holding a substrate;
 - a heater for heating the substrate;
 - an aerosol generator for generating an aerosol comprising droplets of a material solution;
 - a charge applicator for applying a positive or negative charge to the aerosol droplets;
 - a nozzle unit in communication with the aerosol generator for delivering the aerosol to the substrate, the nozzle unit including at least one outlet through which a directed flow of the aerosol is in use delivered and at least one electrode; and
 - a high voltage supply for generating an electric field between the substrate and the at least one electrode such that the directed aerosol flow is in use attracted towards the substrate.
42. The apparatus of paragraph 41, where configured to maintain a decreasing temperature gradient in a direction away from the substrate towards the nozzle unit.
43. The apparatus of paragraph 41 or 42, wherein the at least one electrode extends upstream of the at least one outlet.
44. The apparatus of any of paragraphs 41 to 43, wherein the at least one electrode comprises an elongate element.
45. The apparatus of any of paragraphs 41 to 44, wherein the distal end of the at least one electrode is located at substantially the centre of the at least one outlet.

46. The apparatus of any of paragraphs 41 to 45, wherein the distal end of the at least one electrode includes a single tip.
47. The apparatus of any of paragraphs 41 to 45, wherein the distal end of the at least one electrode includes a plurality of tips.
48. The apparatus of any of paragraphs 41 to 47, wherein the nozzle unit includes a tubular section upstream of each outlet.
49. The apparatus of paragraph 48, wherein the tubular section is an elongate section.
50. The apparatus of paragraph 48 or 49, wherein the tubular section is a linear section.
51. The apparatus of any of paragraphs 48 to 50, wherein the tubular section is substantially cylindrical.
52. The apparatus of any of paragraphs 48 to 51, wherein the at least one electrode extends substantially entirely through the associated tubular section.
53. The apparatus of any of paragraphs 48 to 52, wherein the at least one electrode extends substantially along the central axis of the associated tubular section.
54. The apparatus of any of paragraphs 48 to 53, wherein at least the inner surface of the tubular section is composed of an insulating material.
55. The apparatus of any of paragraphs 41 to 54, further comprising a gas supply unit in communication with the aerosol generator for supplying a flow of a carrier gas for entraining the aerosol and delivering the same through the nozzle unit.
56. The apparatus of any of paragraphs 41 to 55, wherein the at least one outlet is directed upwards, preferably substantially vertically upwards.

00020252

A⁹

57. The apparatus of any of paragraphs 41 to 56, wherein the distance between the at least one outlet and the substrate is less than about 100 mm, preferably less than about 50 mm.
58. The apparatus of any of paragraphs 41 to 57, wherein the nozzle unit and the substrate are held in fixed relation.
59. The apparatus of any of paragraphs 41 to 57, wherein the nozzle unit and the substrate are configured so as to be movable relative to one another.
60. The apparatus of paragraph 59, wherein the substrate is rotatable, tiltable and/or translatable relative to the nozzle unit.
61. The apparatus of any of paragraphs 41 to 60, wherein the nozzle unit includes a perforated member upstream of the at least one outlet.
62. A method of fabricating a powder, preferably an ultrafine powder, comprising the steps of:
providing a heated zone;
generating an aerosol comprising droplets of a material solution;
providing a nozzle unit for delivering the aerosol to the heated zone, the nozzle unit including at least one outlet through which a directed flow of the aerosol is delivered and at least one electrode;
charging the aerosol droplets with a positive or negative charge;
providing a flow of the aerosol through the nozzle unit so as to deliver a directed flow of the aerosol from the at least one outlet; and

generating an electric field between the heated zone and the at least one electrode such that the directed aerosol flow is attracted towards the heated zone where the aerosol droplets react homogeneously in the gas phase to-form a powder.

63. An apparatus for fabricating a powder, preferably an ultrafine powder, comprising:
- a heater for providing a heated zone;
 - an aerosol generator for generating an aerosol comprising droplets of a material solution;
 - a charge applicator for applying a positive or negative charge to the aerosol droplets;
 - a nozzle unit in communication with the aerosol generator for delivering the aerosol to the heated zone, the nozzle unit including at least one outlet through which a directed flow of the aerosol is in use delivered and at least one electrode; and
 - a high voltage supply for generating an electric field between the heated zone and the at least one electrode such that the directed aerosol flow is in use attracted towards the heated zone where the aerosol droplets react homogeneously in the gas phase to form a powder.--

IN THE CLAIMS:

Please amend the claims without prejudice, without admission, without surrender of subject matter, and without any intention of creating any estoppel as to equivalents, as follows: